Ultrashort Laser Pulse Propagation in Water

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LONG-TERM GOALS

The major objectives of this proposal are twofold. The first objective is to perform both an experimental and theoretical study of the factors affecting the propagation distance behavior of ultrashort (femtosecond) laser pulses in water. This study will be conducted in the so-called linear regime which involves laser intensities that are below the threshold where nonlinear effects set in. A fundamental problem, which will be resolved by this research, is whether or not the temporal width and spacing of short pulses affects its absorption spectrum. If one simply knows the spectral composition of the input pulse and the absorption spectrum of the water, is this sufficient to predict the temporal evolution of the pulse? If such is not the case, then we will have to consider the way the system responds to pulse widths and pulse spacing which are short compared to vibrational relaxation times in water.

The second phase of the research will be to explore the nonlinear regime where dramatic changes to the temporal, spatial, and spectral properties of the medium occur. The primary processes being self-focusing and self-phase modulation due to the Kerr effect (also called the quadratic electro-optic effect which was discovered by John Kerr in 1875). Self-focusing can lead to an enormous increase in the peak intensity where long filaments can occur and in some cases lead to supercontinuum generation or "white light" generation first discovered in 1970.

OBJECTIVES

The propagation of an ultrashort pulse of light through a linear and absorptive medium such as water, is of great fundamental importance for several reasons. One of the most important of which is that it may be possible to transmit information over much greater distances using ultrashort pulses compared to propagation distances achieved by using pulses with long time durations, including CW (continuous waves). The first prediction of optical precursors was given by both Sommerfeld and Brillouin[1] in 1914 using an asymptotic method now called steepest descent. Their analysis was based on a step-modulated field propagating through a Lorentz dielectric which is nothing more than a collection of damped harmonic oscillators. Later refinements to their conclusions were made by Oughstun and Sherman[2]. The first measurements which claimed to observe optical precursors in deionized water were made by Choi and Österberg[3] where they found that the precursors were attenuated non-exponentially with distance. What was extremely significant about this work was that they found the

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pulse energy detected at a distance of 5 m in water with a 60 fs pulse was one hundred times greater than the signal obtained using a 900 fs pulse (having the same fundamental wavelength) which had the usual Lambert-Beer behavior (It should be noted that the so called Lambert-Beer law was first discovered by Pierre Bouguer in his published work in 1729 titled *Essai d'optique sur la gradation de la lumiére* and we will hereafter refer to it as the Bouguer-Lambert-Beer Law or BLB. The law is valid for monochromatic sources but unfortunately researchers have mistakenly applied it to pulse propagation where the bandwidth covers changes in the absorption coefficient). Their conclusions were brought into serious doubt by Alfano[4] et al. who claimed that their bandwidth was not wide enough to cross the 760 nm absorption band of water and therefore that their conclusions were questionable.. In a later paper, Fox and Österberg[5] found deviation from BLB when they used pulses with temporal widths of 60 fs and a repetition rate of 1 kHz where they observed more than two orders of magnitude less absorption after propagation through 6 m of water when compared to BLB. They also found that BLB was not violated for pulses with varying bandwidth with temporal widths of 900 fs (pulses centered at 800 nm) and repetition rates of 80 MHz. Li et al[6] have performed a more recent measurement for liquid water and they found deviations from the BLB law after the pulse had propagated only 1.5 m and the deviation continued to increase out to 3.5 m which was as far as they could detect the signal. The shortest pulse they used was 10 fs with a repetition rate of 75 MHz which gives a pulse spacing of 13.3 ns. They showed that deviation from BLB behavior only occurred when the pulse bandwidth was of the order of 100 nm but for shorter bandwidths (< 30 nm) BLB behavior was verified. It is important to note that this pulse spacing is much longer than the vibrational relaxation times in liquid water which are typically of the order of a picosecond. The most recent experiments by Okawachi et al.[7] used four different laser systems centered at 800 and 1530 nm to show the dependence of pulse duration and repetition rate on pulse absorption. Their bandwidths ranged from 20-60 nm and they showed that BLB behavior was obeyed in every case. It is now clear that there is a real enigma associated with the entire phenomena of ultrashort laser propagation in water.

APPROACH

Experimental:

There are three basic improvements over what has been done in the past that we are proposing:

(a) Improved input pulse shapes

The laser system that we will use in this experiment is the one we currently use for femtosecond CARS spectroscopy of dipicolinic acid and bacterial endospores[8-11]. We have an amplified femtosecond laser (Mira + Legend, Coherent) whereby we obtain up to 1 mJ per 35 fs pulse (at 1 kHz repetition rate) at the fundamental 800 nm wavelength. We can control the chirp (positive or negative) of these 800 nm pulses over a wide range (increasing the pulse duration by up to two orders of magnitude, to over 3 ps, while keeping the spectrum unchanged) very easily, and very precisely by adjusting our pulse compressor. Thus we will be able to study propagation of laser pulses of different length but having identical spectra. Fig. 1 shows the experimentally measured pulse shapes obtained by this method and measured by our home-made variation of frequency-resolved optical gating (FROG).

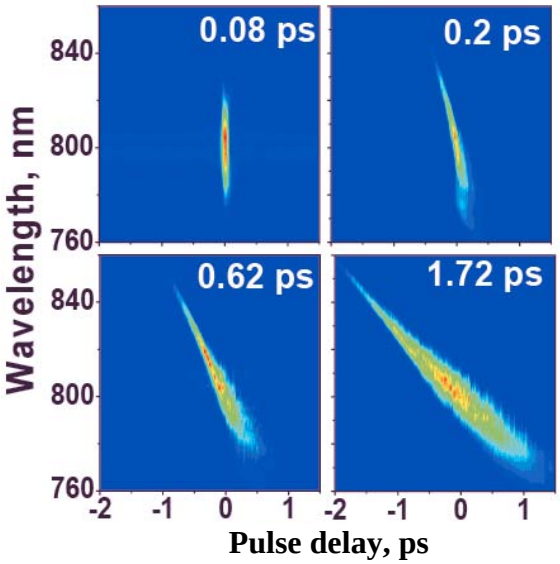

Fig. 1. Chirped pulses obtained by adjusting the pulse compressor and measured by a modified frequency-resolved optical gating technique. The numbers in the upper right corner of each plot give the pulse duration; the spectra of all four pulses are identical.

Alternatively, we can modify the pulse spectral intensities while keeping the spectral phases fixed. Our home-made pulse shaper allows us to make the spectral width up to 100 times narrower and the pulse length correspondingly longer without introducing any chirp. Moreover, our pulse shaper allows us to fine-tune the precise frequency of these narrow-band pulses. We will use such spectrally-modified pulses for the detailed study of light propagation through water. In addition, we have the capability to produce some more complex pulse shapes, should it become necessary or interesting. Our recent paper[12] gives a colorful illustration of potential possibilities, and shows an example of the use of pulse shaping for coherent Raman spectroscopy. More complex pulse shapes will be particularly important for the studies of nonlinear pulse propagation. In the second year of the project, we plan to purchase and use a commercially available computer-programmable pulse shaper for fast (and potentially adaptive) shaping of the laser pulses propagating through water.

Further into the project, we will expand our work into other wavelength regions, and use our two computer-controlled optical parametric amplifiers (OPerA, Coherent), pumped by our amplified femtosecond laser system (described above). Signal and idler pulses obtained from the two OPAs can be frequency-doubled or mixed with the fundamental to produce up to 20 μJ per 50 fs pulse at tunable visible wavelengths. In addition, we may use un-amplified light from the fs oscillator (at 85 MHz), for fast-repetition-rate low-peak-power experiments.

While the previous experimental work on optical precursors suffered from limited choice of laser sources, our laser system will allow great flexibility and precise control over the pulse shapes, for a broad range of well-controlled measurements.

(b) Spectrally-resolved highly-sensitive measurement at the output

We will measure the transmitted spectral intensities (and not just the total output power). We will use a state-of-the-art CCD-equipped spectrometer which will count photons at each wavelength separately. A second CCD-equipped near-infrared spectrometer will allow us to extend our overall working wavelength range. What has been used in many of the past experiments were photomultiplier tubes (PMT's). A PMT is a "bucket" detector that just counts the total number of photons and does not provide spectral resolution (unless it is combined with a spectrometer, in which case a tedious scanning is needed). PMT's are virtually unsurpassed in sensitivity; however, a good cooled CCD can be almost as sensitive. Highly sensitive signal detection over a large dynamic range is essential for the success of the proposed project, since the intensity of light transmitted through many meters of water will vary over many orders of magnitude as a function of propagation length. Our group has a great deal of experience in performing measurements where the signal varies up to over nine decades. As an example, we reproduce here a figure[13] [Fig. 2] which shows signal varying over seven orders of magnitude. That particular set of data was obtained without spectral resolution. Currently we are capable of achieving similar or even higher sensitivity and dynamic range with spectral resolution, which is afforded by the use of a spectrometer equipped with a cooled CCD. Our measurements on pulse propagation through water will be done with spectral resolution.

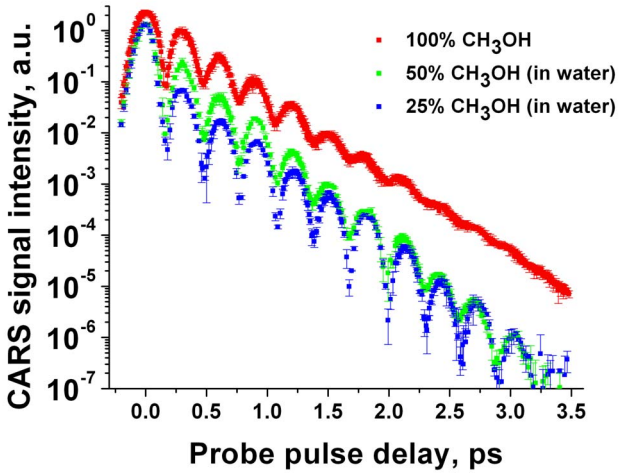

Fig. 2. An example from our previous work, where we used three ultrashort pulses at different wavelength (810 nm, 660 nm, and 575 nm) and measured femtosecond coherent anti-Stokes Raman scattering (CARS) signal (measured in methanol-water solutions) varying in magnitude over many decades[15].

At a further stage of the project we plan to measure the transmitted pulse shapes. For relatively small propagation distances (several meters) we will use a commercial autocorrelator (presently available in our lab), and a FROG apparatus (that we intend to purchase). For large propagation distances this measurement will become challenging since the output power will get very low. For the low-power measurements, we will set up a cross-correlation measurement, such as the one that has produced pictures shown in Fig. 1. Another good option for direct pulse shape measurement is an ultrafast streak camera. We have recently installed and tested such a state-of-the-art streak-camera

(Hamamatsu) in our lab, and we will use it to measure the duration of pulses propagated through over 10 meters of water.

(c) Water cell design

We have designed and already constructed our own water cell. Our focus in designing the new cell was to minimize the number of mirror reflections, and to be able to increase or decrease the path lengths continuously and not just in steps of 0.5 meters.

In previous experiments, the maximum path length through the water was 6 meters. In our water cell, we will be able to run experiments at a maximum path length of 18 meters. A schematic of a simplified experimental set up is provided in Fig. 3. We currently have the equipment installed (as shown) and can have the complete setup within a short period of time.

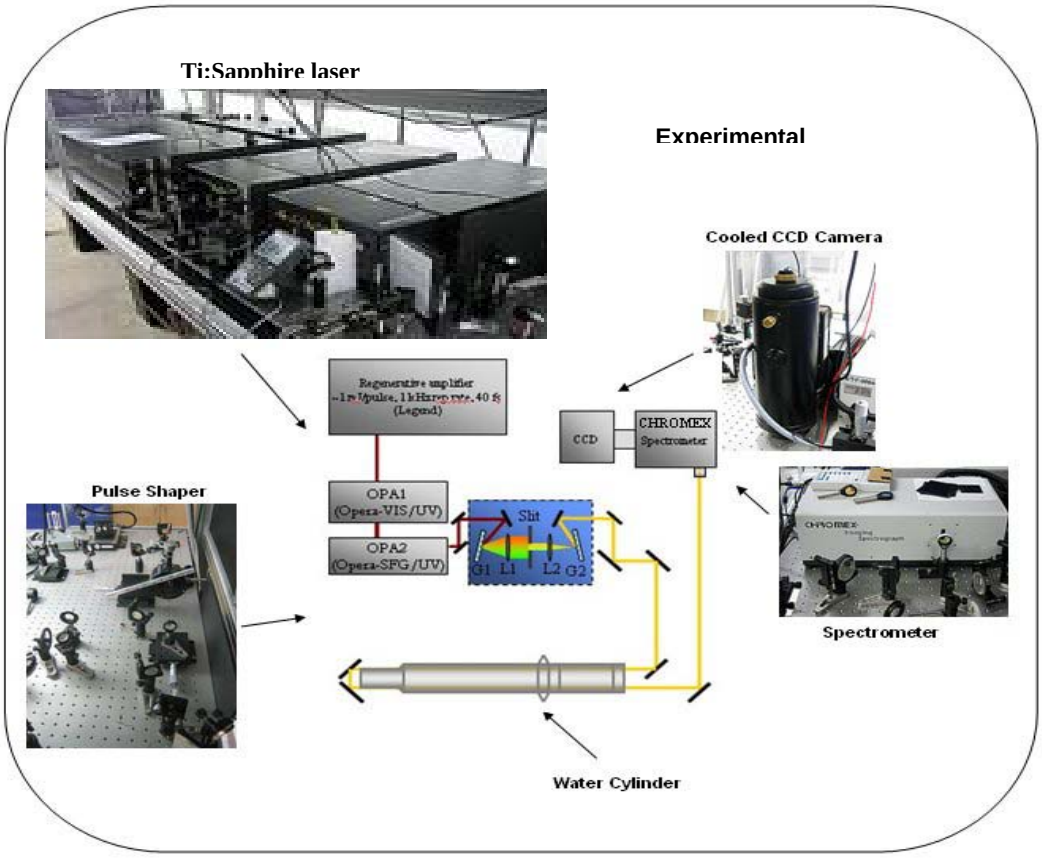

Fig. 3. Experimental setup schematics.

Theoretical:

In order to be able to model the complete pulse propagation process, we must be able to handle the actual pulse shape that will be used in our experiments along with the best data we have on the actual absorption spectra of pure water. In fact the water we will use will be prepared by the same method as in the absorption measurements done by Pope and Fry[14]. Since we have a great deal of experience with the finite difference time domain (FDTD) method, we will have to substantially rewrite parts of it

to handle the actual dispersion of the refractive index of water, particularly around the overtones of the OH-stretch/scissor coupled modes. The regions around the sharp slope changes in refractive index are precisely the regions where the interesting precursors will appear. This type of approach has never been performed but is the *sine qua non* for successful modeling. The basis of our method is to take the two Maxwell curl equations in Rationalized MKSA units and assuming no currents; namely,

$$\begin{aligned} \nabla \times \vec{H} &= \frac{\partial \vec{D}}{\partial t} \\ \nabla \times \vec{E} &= -\frac{\partial \vec{B}}{\partial t} \end{aligned} \tag{1}$$

and then discretize them over a spatial grid. We also need the relation between the displacement current and the electric field; namely, $\vec{D} = \varepsilon(\omega)\vec{E}$ where $\varepsilon(\omega)$ is the frequency dependent permittivity and is related to the refractive index *n* by $n^2 = \varepsilon(\omega)$. In the many papers we have published using the FDTD method to solve scattering from irregularly shaped particles (see references 15-26), we have worked only at a single frequency since the incoming radiation was assumed to be monochromatic. Now when using short pulses, there are a plethora of frequencies involved and so then we must be able to use the actual dispersion relation for the medium involved. In the case of water in the near IR (our pulses are centered around 800 nm) we will use the actual measured absorption data of either Kuo or Segelstein in order to apply the FDTD method to this problem, we need to convert the frequency dependent permittivity to a time dependent one. In particular, the temporal characteristic of the water permittivity should be modeled around the overtones of the OH-stretch/scissor coupled modes. These are the regions where the sharp changes in the slope of the absorption coefficient, denoted by α, occur. Another very interesting aspect of water occurs when it is deuterated to form either HDO or D_2O. These forms will have radically different absorption spectra and we can further test our analyses by studying these different forms of water.

If we denote the refractive index of water by $n=n_R-in_I$ and the permittivity by $\varepsilon=\varepsilon_R-i\varepsilon_I$, where the subscripts R and I denote real and imaginary parts respectively, then it is easy to show that $\varepsilon_R = n_R^2 - n_I^2$ and $\varepsilon_I = 2n_R n_I$. The absorption coefficient α is related to n_I by $\alpha = 4\pi n_I / \lambda_{vac}$ where λ_{vac} denotes the wavelength of the radiation in vacuo.

In order to get the time dependence of water permittivity, we must Fourier transform $\varepsilon(\omega)$. Actually it is more physical to Fourier transform $\varepsilon(\omega)$-1 and to obtain the susceptibility kernel $G(\tau)$ defined by (see Jackson[27]):

$$G(\tau) = \frac{1}{2\pi}\int_{-\infty}^{\infty}[\varepsilon(\omega)-1]e^{-i\omega\tau}d\omega \tag{3}$$

then

$$\begin{aligned} \vec{D}(\vec{x},t) &= \vec{E}(\vec{x},t) + \int_{-\infty}^{\infty} G(\tau)\vec{E}(\vec{x},t-\tau)d\tau \\ &= \vec{E}(\vec{x},t) + \sum_{\tau=0}^{t} G(\tau)\vec{E}(\vec{x},t-\tau)\Delta\tau \end{aligned} \tag{4}$$

Thus we see that the displacement current at a certain instant in time, depends on the electric field from earlier times. It should also be noted that if ε is independent of frequency, then Eq. (4) reduces to $\vec{D} = \varepsilon \vec{E}$ which is what our present FDTD code now uses.

Once we have established the fact that the FDTD method is a viable one for solving this problem, then we would like to next explore the possibility of using the pseudospectral time domain (PSTD) method which we feel will run much faster than the conventional FDTD method. We have tested this method on some canonical scattering problems and found substantial reductions in computation time when compared to the conventional FDTD method. With this method we will actually be able to monitor the complete temporal evolution of the pulse shape.

WORK COMPLETED

a) We have shown that many different types of water have similar absorption curves in the 760-820 nm range.

b) We have shown that broadband continuous and ultrashort laser pulse (around 30 fs pulse duration) light sources experience the same amount of absorption in water samples, so that the absorption curves measured with either source have no significant differences.

c) We have also shown through a very sensitive hole filling technique that ultrashort pulses of duration about 30 fs do not experience any nonlinear attenuation in water until the beam fluence reaches levels great enough to induce intensity-dependent nonlinear effects. Through this technique, we are able to show that the attenuation of ultrashort pulses follows the linear model to within 1% deviation in water.

d) We have now established the fluence level at which nonlinear effects are starting to occur in our water samples; namely, about 210 μJ/cm^2

e) We have studied the feasibility of extending the discontinuous Galerkin (DG) method to a general dielectric medium such as water assisted by the ancillary differential equation (ADE) method. By generalizing the ADE method from the Debye model to a more general case, we concluded that it is feasible to apply the DG method to a general medium.

f) We studied the dynamics of ultrashort pulses in a Lorentz medium, the so-called generalized Sommerfeld and Brillouin precursor method. We were able to show, for the first time, that the saddle point method can be used to analyze ultrashort pulse propagation in water. We showed that the saddle point method is more efficient and faster than the direct integration method we earlier used to study one-dimensional pulse propagation over macroscopic distances (that is, compared with the wavelength) in a general dielectric medium. By applying the saddle point method, we were also able to show that the group velocity dispersion (GVD) approximation for pulse propagation in water is valid for propagation distances up to 6 m.

RESULTS

Here we present the results obtained with the “hole filling” technique that sensitively monitors spectral changes in a laser pulse undergoing propagation in a medium. Output spectra were measured as a function of the input laser power. Figs. 4 (a), (b), and (c) show the input spectra at different input powers, while Figs. 4(d), (e), and (f) show the corresponding output spectra measured with the Ocean

Optics spectrometer. To compare the shapes of the spectra for different input powers we first took the data and divided each measured spectral curve by the corresponding input power. This procedure by itself produced curves deviating from each other by no more than the relative uncertainty of our power measurements (with the absolute experimental uncertainty of 0.1 mW, the relative uncertainty varies from 10% for 1 mW of input power to 0.8% for 12 mW). To avoid these variations due to uncertainty in the power measurements, we normalized each curve to its peak value, in order to be able to look at possible small (well less than 1%) relative variations in the resultant spectral shapes. The insets shown in Figs. 4 (d), (e), and (f) illustrate a mismatch between the different curves at the wavelengths corresponding to the spectral holes. This mismatch is less than 0.1% of the peak value, and is within the spectrometer noise.

The different hole positions were chosen to check the absorption dynamics of the broadband pulses in water. These results show that the difference in the input and output spectra is due only to the linear absorption of water, which has a strong absorption band at around 745-780 nm[28]. In the case of nonlinear spectral transformations that generate new frequencies, the depth of the spectral holes should decrease (hole filling). However, the above results show that the depth of the hole remains the same, which indicates that there was no "hole filling" effect and no new frequencies were generated. In addition, our estimates for the threshold power for nonlinear effects such as self-focusing and self-phase modulation show that in the experiments with the oscillator pulses, we are well below these threshold power ranges.

Fig. 5 shows the measured transmitted spectral intensities at three wavelengths as a function of the input power. The three wavelengths correspond to the low-power spectral hole center (794 nm) and two wavelengths at the spectral wings (758 and 836 nm) having, at low input powers, the same spectral intensity as the hole center. At low input powers (below about 40 mW), we observe linear growth of the spectral intensities, turning to nonlinear behavior at higher input powers.

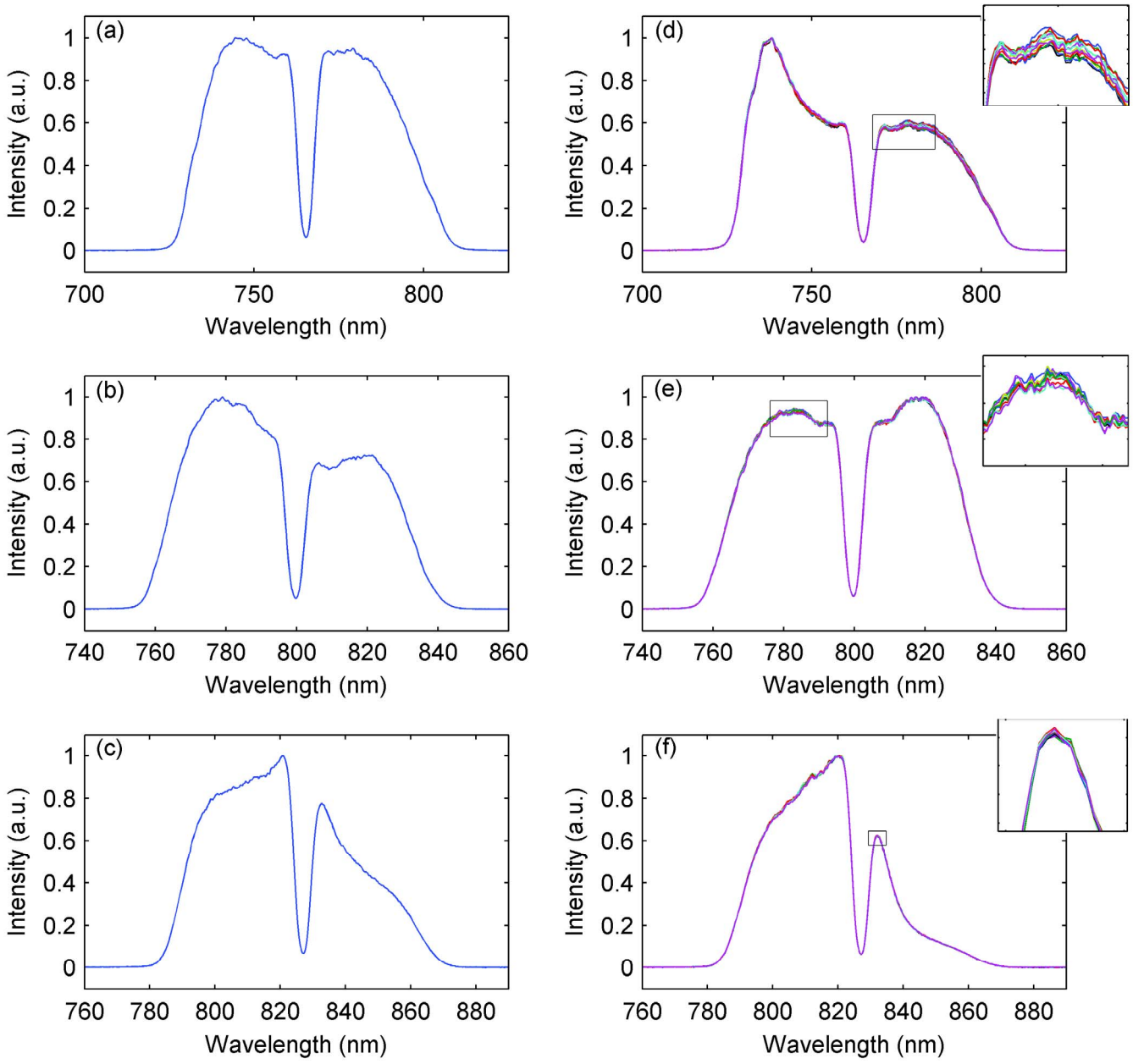

Fig. 4 Results from the spectral hole filling experiment with oscillator 7 fs laser pulses: (a), (b), (c) show the input spectra with different positions of the center of the spectral hole at three different wavelengths 767 nm, 800 nm, and 827 nm.; (d), (e), (f) show the corresponding output spectra after propagation of the pulses through the cell with distilled water.

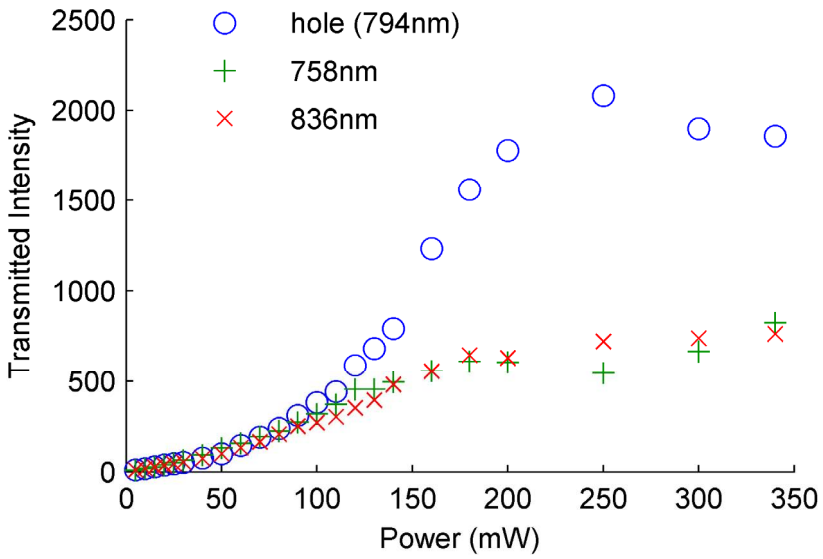

Fig. 5 Transmitted intensity for three wavelengths as a function of beam input power. The wavelength of 794 nm corresponds to the hole in the spectrum. The wavelengths of 758 and 836 nm correspond to the wavelengths on the wings with the spectral intensities equal to the intensity of the hole at the bottom for low beam input powers.

IMPACT/APPLICATION

The work we are doing will definitely lead to a better understanding of the mechanisms responsible for the absorptive properties of water. It may also open a whole new area of research to find out whether pulse duration or pulse repetition rate or pulse power will allow us to propagate signals further in water than using conventional CW systems

TRANSITIONS

If the results of this research uncover novel ways to propagate signals further in water than CW systems, it will have far reaching applications in biomedical research on human tissue since the majority of the human body is water.
Also the techniques we have developed will have definite applications for remote sensing of ocean water and on signal transmission within the water.

RELATED PROJECTS

We use the results from our research on the RaDyO project to give us the IOP's for water we use in our analysis for short pulses.

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1. L.M. Naveira, B.D. Strycker, J. Wang, G.O. Ariunbold, A.V. Sokolov, and G.W. Kattawar, "Propagation of femtosecond laser pulses through water in the linear absorption regime," Appl. Opt. 48, 10, 1828-1836 (2009)[published,refereed]

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